Ferromagnetic to Antiferromagnetic Transition in Fe_xCr_{1-x} Films with Composition: A Transmission MCD Study

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INTRODUCTION

The ability to study element-resolved magnetism using magnetic circular dichroism (MCD) and other soft x-ray magneto-optical techniques enables a variety of studies of both homogeneous and inhomogeneous magnetic thin film systems. The goal of this study is to better understand the transition from ferromagnetism to antiferromagnetism with composition in the Fe_xCr_{1-x} alloy system. This system is especially interesting for reasons both fundamental and practical. Ferromagnetic Fe and antiferromagnetic Cr both take the bcc crystal structure with less than one percent difference in lattice constant, so that the magnetic transition with X at first glance might be thought to involve purely electronic rather than structural effects. This system forms one boundary of the Slater-Pauling curve representing the evolution of magnetic moments with d electron concentration in 3d transition metal alloys, and has been a prototypical system for studying magnetic and structural correlations in alloys with neutron [1,2] and x-ray scattering [3] techniques. More recently, magnetic ordering of Cr monolayers on Fe has been much studied [4,5], as have Fe/Cr multilayer [6,7] and granular alloy [8] films for their magneto-resistive properties. Even with this extensive background on FeCr alloys and related systems, alloy thin films have not been the subject of systematic element-resolved MCD studies that should offer new insight into these systems.

Polycrystalline alloy films ranging from 55 - 75 nm thick were grown by magnetron co-deposition onto silicon nitride membrane substrates and capped with several nm of silicon carbide to prevent oxidation. Transmission MCD measurements were made with magnetization saturated in-plane and circular polarization from out-of-plane radiation on bending magnet beamlines 9.3.2 and 6.3.2. The use of transmission measurements stems from our interest to study the bulk of thin film specimens rather than their near-surface regions.

RESULTS AND DISCUSSION

Thickness effects in transmission x-ray absorption spectra in the vicinity of the sharp white lines manifest themselves as apparent reductions in absorption at white lines, or equivalently as enhanced transmission [9]. This results from unwanted spectral components in the incident beam that fall outside the fundamental energy resolution to enhance apparent transmission at the white line when transmission of the fundamental becomes comparable to that of the spectral impurities. Thickness effects become more problematic as the optical thickness of samples increases, and can be safely ignored only for samples which are optically thin at white line peaks. We made absorption measurements vs. film thickness in pure Fe films (having 1/e length of only 17 nm at the L_3 line) revealing significant thickness effects that varied with the grating used in the monochromator and hence from beamline to beamline. We attribute the thickness effects in the soft x-ray primarily to higher order contamination in the incident beam originating from the gratings. Since MCD signals are the difference of two white line intensities that are distorted differently by thickness effects, great care must be exercised in attempting to obtain quantitative MCD results (sum rules, etc.) from transmission spectra. Since not all samples studied here were optically thin at the Fe and Cr L_2 and L_3 edges, we do not attempt to quantify elemental moments based on these
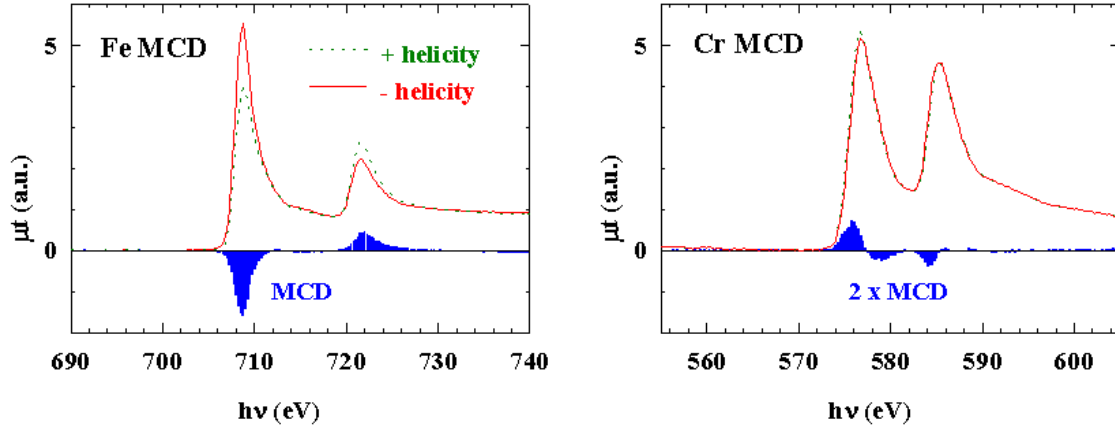
Figure 1. Transmission absorption spectra for a saturated $Fe_{0.85}Cr_{0.15}$ film taken with opposite helicity and the difference yielding the MCD are shown across the Fe (left) and Cr (right) $L_{2,3}$ edges. Cr has a net moment oriented opposite to that of Fe, and a very different spin polarization from that of Fe.

spectra. However, we do believe that the qualitative trends in elemental moments are generally correct. In the conclusions we indicate how we plan to avoid thickness effects in the future.

Absorption and MCD spectra for an alloy film with X = 0.85 are shown at the Fe and Cr $L_{2,3}$ edges in Figure 1. Absorption spectra taken with opposite helicity relative to the magnetization are each normalized to have edge jump of 1 and then subtracted to give the MCD signal. The Fe results are typical of elemental Fe, with strong MCD peaks of opposite signs at the L_3 and L_2 peaks. The Cr MCD spectrum is very different from that of Fe, with a bipolar feature at the L_3 edge and a weaker feature of opposite sign at the L_2 edge. The different shapes of the Fe and Cr MCD are qualitatively understood by realizing that Cr has a lower d-band occupancy than Fe of roughly 2 electrons, and Fe has a roughly half-filled d band. The strong exchange splitting in Fe results in nearly complete spin-polarization characterized by a nearly full spin up (majority) band and nearly empty spin down (minority) band. The absorption is dominated by transitions into the minority band. The spin polarization in Cr is very different from Fe, and can be interpreted as containing empty states of both spins with different energy dependencies. The striking differences between Fe and Cr MCD spectra reveal that a theoretical description in terms of a single spin-polarized band structure describing both Fe and Cr spin polarizations is not possible.

Just the MCD spectra at both the Fe and Cr $L_{2,3}$ edges are shown in Figure 2, and reveal three distinct magnetization regions as a function of composition. In the region from pure Fe to roughly X = 0.58 Fe retains its distinct MCD spectrum. The apparent increase in Fe MCD intensity may indicate a real increase in Fe moment, or may be the result of thickness effects.
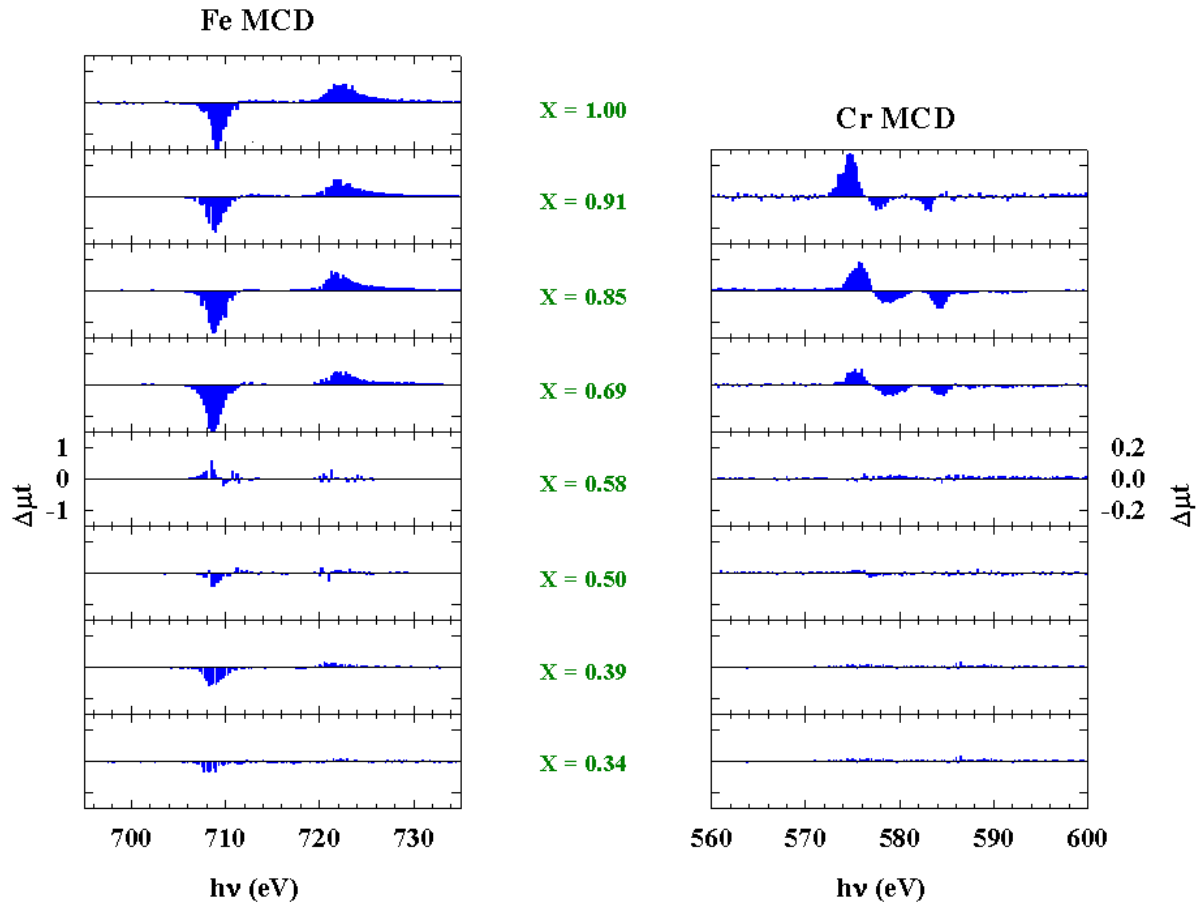
Figure 2. Fe (left) and Cr (right) MCD spectra for a range of alloy compositions reveal three distinct ranges of magnetization as discussed in the text.

The Cr MCD spectra in this range resembles that in Figure 1 but with decreasing intensity as Cr content increases. Evidently at the lowest Cr concentrations Cr atoms have predominantly Fe neighbors, and order ferrimagnetically with respect to the Fe. As more Cr is added in this range, the decreasing Cr moment can be interpreted assuming that Cr is segregating. In this case Cr atoms adjacent to Fe will tend to have moments opposite to those of Fe, while Cr atoms adjacent to Cr in clusters will order antiferromagnetically with respect to the adjacent Cr, resulting in a decrease in net Cr moment. Such a chemical short range order inversion in bulk FeCr alloys has been reported based on neutron scattering [2].

The second region is from $0.58 > X > 0.50$, in which neither Fe or Cr exhibit a significant moment. This composition range corresponds closely to that of a slow-forming σ phase in the equilibrium phase diagram for bulk alloys, although this phase has never been reported in thin films. X-ray diffraction results of films in this region reveal a shift in the 110 Bragg peak to higher angles, indicating a structural distortion characterized by an increased interatomic spacing accompanies the loss of magnetism in this composition range. This may be evidence of incipient σ phase formation, and at least indicates a strong link between structure and magnetism in this system, possibly related in to the invar transition in other binary transition metal alloys. Further experiments will determine if the films in this intermediate composition range are paramagnetic or possibly antiferromagnetic with Fe and Cr sharing the two sublattices.

The third region comprises the Cr-rich samples, and shows the re-emergence of an Fe moment that then disappears with increasing Cr content and no clear Cr moment. This behavior can be interpreted as representing a ferrimagnetic arrangement in which the Fe moments ultimately vanish as the antiferromagnetism of Cr dominates the magnetic behavior.

CONCLUSIONS

This transmission MCD study of Fe_XCr_{1-X} alloys reveals qualitative changes in magnteism with composition that are similar to but somewhat different than those reported in bulk alloys. The element-resolved sensitivity of the x-ray techniques provides information in all three composition ranges that is not possible by most techniques, and the L edge spectroscopies provide very direct information on the changing average spin polarization around Fe and Cr atoms with composition. The evident thickness effect that precludes determination of absolute moments and their resolution into spin and orbital moments for these transmission measurements will be overcome by future measurements of the Faraday rotation spectra, which are free from thickness effects and can also yield the moment information via Kramers-Kronig transformation analysis [10]. Several other studies using soft x-rays suggest themselves based on these early results, including a diffuse scattering study of the alloy films especially in the Fe-rich region where chemical segregation is suspected. Tuning to the Fe and Cr edges will significantly enhance diffuse small angle scattering [11] from such segregation if it exists.

REFERENCES

1. C.G. Shull and M.G. Wilkinson, Phys. Rev. **97**, 304 (1955).
2. I. Mirebeau, M. Hennion and G. Parette, Phys. Rev. Lett. **53**, 687 (1984).
3. L. Reinhard, J.L. Robertosn, S.C. Moss, G.E. Ice, P. Zschack and C.J. Sparks, Phys. Rev. B **45**, 2662 (1992).
4. L.E. Klebanoff, S.W. Robey, G. Liu, and D.A. Shirley, Phys. Rev. B **30**, 1048 (1984).
5. A. Davies, J.A. Stroscio, D.T. Pierce, and R.J. Celotta, Phys. Rev. Lett. **76**, 4175 (1996).
6. S.S.P. Parkin, N. More and K.P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
7. E.E. Fullerton, S.D. Bader and J.L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).
8. K. Takanashi, T. Sugawara, K. Hono and H. Fujimori, J. Appl. Phys. **76,** 6790 (1994).
9. L.G. Parratt, C.F. Hempstead, and E.L. Jossem, Phys. Rev. **105**, 1228 (1957).
10. J.B. Kortright, M. Rice and R. Carr, Phys. Rev. B **51**, 10240 (1995).
11. Walton and J.B. Kortright, this compendium, and C.C. Walton, Ph.D. Thesis U.C. Berkeley and LBNL Report LBNL-41425 (December, 1997).